



THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of  
PLANTENGA, F. et al.

Group Art Unit: 1764  
Examiner: N. G. Preisch  
Docket No.: ACH 2696US

Application No.: 09/540400  
Filed: March 31, 2000

For: PROCESS FOR EFFECTING ULTRA-DEEP HDS OF HYDROCARBON  
FEEDSTOCKS

Declaration under 37 C.F.R. §1.132

Sir:

I, Franciscus Wilhelmus van Houtert, a citizen of the Netherlands, hereby declare and state:

1. I have a degree in Chemical Engineering which was conferred upon me by the Technical University in Eindhoven in 1980.
2. I have been employed by Akzo Nobel since 1980 and I have a total of 22 years of work and research experience in the field of cracking catalysts and hydrotreating catalysts. I am presently employed as Section Head Application Research Hydrotreating Catalysts.
3. I am one of the inventors of the present application.

I herewith declare as follows:

Under my supervision, the following tests were carried out:

A first catalyst containing nickel, molybdenum, and an organic additive on a carrier and a second catalyst containing cobalt, molybdenum, and an organic additive on a carrier were tested in sulfur removal from hydrocarbon feeds in two experiments with different sulfur contents of the feed and product.

The testing took place in an upflow tubular reactor. Each reactor tube contained 75 ml of catalyst homogeneously intermixed with 70 ml of carborundum particles.

#### First experiment

The feed applied was a diesel feedstock with the following properties.

|                                |           |
|--------------------------------|-----------|
| Nitrogen (ASTM D-4629) (ppmwt) | 338       |
| Sulfur (ASTM D-4294) (ppmwt)   | 13891     |
| Density 15°C (g/ml)            | 0.8661    |
| Dist. (°C)                     | ASTM D-86 |
| IBP                            | 178       |
| 5 vol. %                       | -         |
| 10 vol. %                      | 217       |
| 30 vol. %                      | 251       |
| 50 vol. %                      | 279       |
| 70 vol. %                      | 307       |
| 90 vol. %                      | 348       |
| 95 vol. %                      | 365       |
| FBP                            | 377       |

The catalysts were sulfided using an SRLGO in which dimethyl disulfide had been dissolved to a total S content of 2.5 wt. %.

The catalysts were tested under the following conditions:

|                              |         |
|------------------------------|---------|
| temperature                  | 350°C   |
| H <sub>2</sub> / oil ratio   | 200 n/l |
| reaction pressure            | 30Bar   |
| liquid hourly space velocity | 2.00    |

The products from the different runs were analysed. The results thereof are given below.

#### Sulfur analysis of products obtained

|   | ppm S in the product<br>Ni-Mo additive catalyst | ppm S in the product<br>Co-Mo additive catalyst |
|---|---|---|
| 1 | 352   | 217   |

It appears that in this experiment, which is outside the scope of the presently claimed invention, the cobalt-containing catalyst shows a much higher activity than the nickel-containing catalyst under the same conditions.

Second experiment – ultradeep-HDS

The feed applied was a diesel feedstock with the following properties.

|                                |           |
|--------------------------------|-----------|
| Nitrogen (ASTM D-4629) (ppmwt) | 86        |
| Sulfur (ASTM D-4294) (ppmwt)   | 12408     |
| Density 15°C (g/ml)            | 0.8371    |
| Dist. (°C)                     | ASTM D-86 |
| IBP                            | 184       |
| 5 vol. %                       | 219       |
| 10 vol. %                      | 231       |
| 30 vol. %                      | 265       |
| 50 vol. %                      | 287       |
| 70 vol. %                      | 310       |
| 90 vol. %                      | 345       |
| 95 vol. %                      | 360       |
| FBP                            | 374       |

The catalysts were sulfided using an SRLGO in which dimethyl disulfide had been dissolved to a total S content of 2.5 wt. %.

The catalysts were tested under the following conditions:

|                              |      |
|------------------------------|------|
| temperature                  | 340  |
| H <sub>2</sub> / oil ratio   | 200  |
| reaction pressure            | 45   |
| liquid hourly space velocity | 2.50 |

The products from the different runs were analysed. The results thereof are given below.

Sulfur analysis of products obtained

|   | ppm S in the product<br>Ni-Mo additive catalyst | ppm S in the product<br>Co-Mo additive catalyst |
|---|---|---|
| 1 | 10  | 67  |

It appears that for ultra-deep HDS the nickel-containing catalyst shows a much higher activity than the cobalt-containing catalyst under the same conditions.

I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and

the like so made are punishable by fine and/or imprisonment under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing therefrom.

Date:

*March 25, 2003*

A handwritten signature in black ink, appearing to read 'F.W. van Houtert', with a long horizontal line extending from the bottom of the signature.

F.W. van Houtert